

Amendments to the Specification:

On page 1, after the title, insert the following:

CROSS-REFERENCE TO RELATED APPLICATION

This application is the U.S. national phase of PCT Appln. No. PCT/EP2004/012201 filed October 28, 2004, which claims priority to German application 103 51 803.7 filed November 6, 2003.

BACKGROUND OF THE INVENTION

1. Field of the Invention

Please amend the paragraph beginning on page 1, line 4, as shown below:

The invention relates to a method for the production of phosphonic ester-modified organosilicon compounds by reaction of silanes ~~contained~~ containing phosphonic ester groups with reactive silicon compounds.

On page 1, before the paragraph beginning on line 9, please add the following:

2. Description of the Related Art

Please amend the paragraph on page 1, line 9, as shown below:

Phosphonic ester-modified ~~silicons~~ organosilicon compounds are of great economic interest for a multitude of sectors. For example, they may be used as lubricants on metals and textiles, flame-retardant additives, adhesion promoters, additives for cosmetics or laundry detergents,

defoamers, release agents, damping fluids, heat transfer fluids, antistatic agents or for polishes and coatings.

Please amend the paragraph on page 1, line 18, as shown below:

Phosphorus-modified siloxanes are prepared generally by reaction of trialkyl phosphites with chloropropyl-modified siloxanes, as described, for example in Gallagher et al., J. Polym. Sci. Part A, Vol. 41, 48-59 (2003). Unfortunately, long reaction times and high temperatures are needed for this reaction, which leads to rearrangements in the product and thus to yield losses and also to unwanted by-products.

Please amend the paragraph on page 1, line 27, as shown below:

The reaction of trialkyl phosphites with chloromethyl-modified siloxanes as described in U.S. patent [[US]] 2,768,193 or by Gallagher et al. proceeds significantly more quickly but has the disadvantage that the siloxanes thus produced are difficult to purify by distillation on account of their high boiling point. In addition, however, the progress of this reaction is slow, since the concentration ~~with~~ of the reactive groups is greatly reduced by dilution with unreactive dimethylsiloxo units, resulting in reaction times in the region of several hours.

On page 2, before line 1, please insert the following heading:

SUMMARY OF THE INVENTION

Please amend the paragraph on page 2, line 1, as shown below:

[[The]] An object ~~on which~~ of the invention, ~~is based~~, then, was to provide a method for the production of phosphonic ester-modified organosiloxanes that makes it possible, starting from commercially available chemicals, to produce [[the]] phosphonic ester-modified organosiloxanes extremely simply, with short reaction times, and in high yields. These and

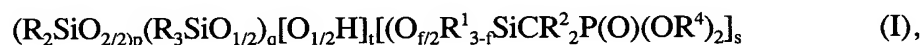
other objects are attained by reaction of an alkoxysilane phosphonic ester with a silanol-functional organosilicon compound.

On page 2, before line 9 , please insert the following heading:

DETAILED DESCRIPTION OF THE PREFERRED EMBODIMENTS

Please amend the paragraph beginning on page 2, line 9, as shown below:

The invention provides a method for the production of phosphonic ester-modified organosiloxanes of the general formula



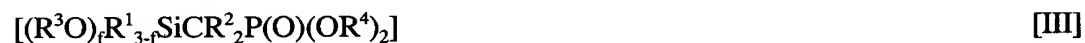
in which

- R** is a hydrogen atom or a monovalent, optionally -CN-, -NCO-, NR^5_2 -, -COOH-, -COOR⁵-, -halogen-, -acryloyl-, -epoxy-, -SH-, -OH- or -CONR⁵₂- substituted Si-C-bonded C₁-C₂₀ hydrocarbon radical or C₁-C₁₅ hydrocarbonoxy radical in which one or more nonadjacent methylene units in each case may be replaced by groups -O-, -CO-, -COO-, -OCO- or -OCOO-, -S- or -NR⁵- and in which one or more nonadjacent methine units may be replaced by groups[[,]] -N=, -N=- or -P=,
- R¹** is a hydrogen atom or a monovalent, optionally -CN-, -NCO-, -COOH-, -COOR⁵-, -halogen-, -acryloyl-, -SH-, -OH- or -CONR⁵₂- substituted Si-C-bonded C₁-C₂₀ hydrocarbon radical or C₁-C₁₅ hydrocarbonoxy radical in which one or more nonadjacent methylene units in each case may be replaced by groups -O-, -CO-, -COO-, -OCO-, or -OCOO-, -S-, or -NR⁵- and in which one or more nonadjacent methine units may be replaced by groups[[,]] -N=, -N=- or -P=,
- R²** is hydrogen or an optionally -CN- or halogen-substituted C₁-C₂₀ hydrocarbon radical,
- R⁴** is hydrogen or an optionally -CN- or halogen-substituted C₁-C₂₀ hydrocarbon radical or substituted or unsubstituted polyalkylene oxides having 1 to 4000 carbon atoms,

R^5 is hydrogen or an optionally -CN- or halogen-substituted C_1 - C_{10} hydrocarbon radical,
 p is 0 or an integer of from 1 to 100 000,
 q is 0 or an integer of from 1 to 100 000,
 f is the number 1 or 2 or 3,
 s is an integer which is at least 1 and
 t is 0 or an integer which is at least 1,
 $p+q$ being an integer which is at least 1,

characterized in that

at least one silane of the formula



is reacted with at least one silicon compound of the general formula



where

R^3 is hydrogen or an optionally -CN- or halogen-atom-substituted C_1 - C_{20} hydrocarbon radical, and
 m is an integer 1 or 2,
 R , R^1 , R^2 , R^4 , p , q , f and s have the above definitions.

Please amend the paragraph beginning on page 3, line 31, as shown below:

The phosphonic ester-modified organosiloxanes of the general formula (I) have a phosphonic ester function which is attached via a C atom through a Si-C-P bond to a silicon atom of the organosilicon[[e]] compound.

Please amend the paragraphs beginning on pages 3 and 4, lines 36 and 5 as shown below:

The radicals **R** may be alike or different, substituted or unsubstituted, aliphatically saturated or unsaturated, aromatic, cyclic, straight-chain or branched. **R** preferably has 1 to 12 carbon atoms, in particular 1 to 6 carbon atoms, and is preferably unsubstituted. More ~~[[P]]~~preferably, **R** is a straight chain or branched C₁-C₆ alkyl radical, the methyl, ethyl, phenyl, vinyl and trifluoropropyl radicals being particularly preferred.

Please amend the paragraph beginning on page 4, line 9, as shown below:

The radicals **R**¹ may be alike or different, substituted or unsubstituted, aliphatically saturated or unsaturated, aromatic, cyclic, straight-chain or branched. **R**¹ is preferably a C₁-C₁₀ alkyl radical or phenyl radical, especially a branched or unbranched C₁-C₃ alkyl radical, which may also be substituted. With particular preference, **R**¹ is a methyl radical or ethyl radical.

Please amend the paragraph beginning on page 4, line 18, as shown below:

The radicals **R**² may, independently of one another, likewise be substituted or unsubstituted, aliphatically saturated or unsaturated, aromatic, cyclic, straight-chain or branched. **R**² is preferably a C₁-C₃ alkyl radical or hydrogen atom. With particular preference, **R**² is hydrogen atom.

Please amend the paragraph beginning on page 4, line 25, as shown below:

The radicals **R**³ may, independently of one another, likewise be substituted or unsubstituted, aliphatically saturated or unsaturated, aromatic, cyclic, straight-chain or branched. **R**³ is preferably a C₁-C₅ alkyl radical or hydrogen atom, especially C₁-C₃ alkyl radical or hydrogen atom. With particular preference, **R**³ is a methyl or ethyl radical.

Please amend the paragraph beginning on page 4, line 33, as shown below:

The radicals R^4 may independently of one another, likewise be substituted or unsubstituted, aliphatically saturated or unsaturated, aromatic, cyclic, straight-chain or branched. R^4 is preferably a C_1 - C_{12} alkyl or aryl radical. With particular preference, R^4 is a methyl, ethyl, butyl, phenyl or cyclohexyl radical. R^4 may, if desired, also contain heteroatoms such as, ~~for example,~~ an oxygen atom or nitrogen atom or other functional group[[s]].

Please amend the paragraph beginning on page 5, line 5, as shown below:

The radicals R^5 are preferably a hydrogen atom or an unsubstituted or substituted C_1 - C_{10} alkyl radical.

Please amend the paragraph beginning on page 6, line 23, as shown below:

Preferably the silicon compounds of the formula (IV) that are used in accordance with the invention are $[(H_3C)_2SiO_{2/2}]_{15}[O_{1/2}H]_2$, $[(H_3C)_2SiO_{2/2}]_{30}[O_{1/2}H]_2$, $[(H_3C)_2SiO_{2/2}]_{55}[O_{1/2}H]_2$, ~~$[(H_3C)_3SiO_{2/2}]_{15}[O_{1/2}H]_2$, $[(H_3C)_2SiO_{2/2}]_{150}[O_{1/2}H]_2$, $[(H_3C)_3SiO_{1/2}][(H_3C)_2SiO_{2/2}]_{20}[O_{1/2}H]$ and $[(H_3C)_3SiO_{1/2}][(H_3C)_2SiO_{2/2}]_{50}[O_{1/2}H]$, with $[(H_3C)_2SiO_{2/2}]_{15}[O_{1/2}H]_2$, $[(H_3C)_2SiO_{2/2}]_{30}[O_{1/2}H]_2$, and $[(H_3C)_2SiO_{2/2}]_{55}[O_{1/2}H]_2$, being particularly preferred.~~

Please amend the paragraph beginning on page 7, line 5, as shown below:

In the process of the invention silanes of the general formula (III) are reacted with silicon compounds of the general formula (IV). This reaction may take place under mild conditions, which allows reaction of an Si-OH group on the siloxane of the general formula (IV) with the silane of the general formula (III) without Si-O-Si bonds of the siloxanes of the formula (IV) being broken and possibly re-established. In this case it is advantageously possible ~~advantageously in some cases~~ to do without the use of special catalysts in some cases. However, the reaction according to the invention progresses with the use of catalysts, particularly those which are used in accordance with the prior art for preparing alkoxy-

terminated siloxanes or for accelerating the reaction of alkoxysilanes, in RTV-1 compositions, for example, more rapidly and more completely. It is, however, also possible to use other catalysts, such as phosphoric acids or partial phosphoric esters, such as isopropyl phosphate, for example.

Please amend the paragraph beginning on page 12, line 11, as shown below:

A 500 ml three-necked flask with dropping funnel and reflux condenser was charged under nitrogen atmosphere with 26.1 g of diethoxyphosphitomethyldimethoxymethylsilane (0.10 mol, GC 98%), whose preparation is described above under "Silane A". Following the addition of 0.5% by weight of isopropyl phosphate catalyst and heating to 60°C, 220 g of a doubly OH-terminated polydimethylsiloxane (M = 1100 g/mol; 0.2 mol) were slowly added dropwise over the course of 10 minutes with vigorous stirring. Subsequently the reaction mixture was heated at 80°C for 120 minutes. After the alcohol formed has been stripped off under reduced pressure, 239 g of poly((diethoxyphosphitomethyl)methylsiloxane-co-dimethylsiloxane) were obtained with an average molecular weight (number average) of 2500 g/mol. This is a linear block copolymer in which two polydimethylsiloxane chains are joined via a diethoxyphosphitomethyl[[]]methylsiloxane unit.

Please amend the paragraph beginning on page 12, line 33, as shown below:

A 250 ml three-necked flask with dropping funnel and reflux condenser was charged under nitrogen atmosphere with 48.9 g of diethoxyphosphitomethyldimethylmethoxysilane (0.20 mol, GC 98%), whose preparation is described above under "Silane B". Following the addition of 0.5% by weight of isopropyl phosphate catalyst and heating to 60°C, 110 g of a doubly OH-terminated polydimethylsiloxane (M = 1100 g/mol; 0.1 mol) were slowly added dropwise over the course of 10 minutes with vigorous stirring. Subsequently the reaction mixture was heated at 80°C for 120 minutes. After the alcohol formed has been stripped off under reduced pressure, 153 g of a polydimethylsiloxane containing diethoxyphosphitomethyl end groups were obtained with an average molecular weight (number average) of 1500 g/mol. This is a linear block copolymer in which a diethoxyphosphitomethyl[[]]methylsiloxane unit is joined to each of the two ends of a polydimethylsiloxane chain.

Please amend the paragraph beginning on page 13, line 17, as shown below:

A 500 ml three-necked flask with dropping funnel and reflux condenser was charged under nitrogen atmosphere with 48.9 g of diethoxyphosphitomethyldimethylmethoxysilane (0.20 mol, GC 98%), whose preparation is described above under "Silane B". Following the addition of 0.5% by weight of isopropyl phosphate catalyst and heating to 60°C, 300 g of a doubly OH-terminated polydimethylsiloxane (M=3000 g/mol; 0.1 mol) were slowly added dropwise over the course of 10 minutes with vigorous stirring. Subsequently the reaction mixture was heated at 80°C for 240 minutes. After the alcohol formed has been stripped off under reduced pressure, 343 g of a polydimethylsiloxane containing diethoxyphosphitomethyl end groups were obtained with an average molecular weight (GPC, number average) of 3600 g/mol. This is a linear block copolymer in which a diethoxyphosphitomethyl[D]]methylsiloxane unit is joined to each of the two ends of a polydimethylsiloxane chain.

Please amend the paragraph beginning on page 14, line 3, as shown below:

A 250 ml three-necked flask with dropping funnel and reflux condenser was charged under nitrogen atmosphere with 4.9 g of diethoxyphosphitomethyldimethylmethoxysilane (0.02 mol, GC 98%), whose preparation is described above under "Silane B". Following the addition of 0.5% by weight of isopropyl phosphate catalyst and heating to 60°C, 108 g of a doubly OH-terminated polydimethylsiloxane (M=10 800 g/mol; 0.01 mol) were slowly added dropwise over the course of 10 minutes with vigorous stirring. Subsequently the reaction mixture was heated at 80°C for 300 minutes. After the alcohol formed has been stripped off under reduced pressure, 110 g of a polydimethylsiloxane containing diethoxyphosphitomethyl end groups were obtained with an average molecular weight (GPC, number average) of 12 300 g/mol. This is a linear block copolymer in which a diethoxyphosphitomethyl[D]]methylsiloxane unit is joined to each of the two ends of a polydimethylsiloxane chain.

Please amend the paragraph beginning on page 14, line 25, as shown below:

A 250 ml three-necked flask with dropping funnel and reflux condenser was charged under nitrogen atmosphere with 26.1 g of diethoxyphosphitomethyldimethoxymethylsilane (0.10 mol, GC 98 %), whose preparation is described above under "Silane B". Following the addition of 0.5% by weight of isopropyl phosphate catalyst and heating to 60°C, 110 g of a doubly OH-terminated polydimethylsiloxane (M = 1100 g/mol; 0.1 mol) were slowly added dropwise over the course of 10 minutes with vigorous stirring. Subsequently the reaction mixture was heated at 80°C for 120 minutes. After the alcohol formed has been stripped off under reduced pressure, 121 g of a polydimethylsiloxane containing diethoxyphosphitomethyl end groups and were obtained with an average molecular weight (number average) of 10 060 g/mol. This is a linear block copolymer in which a diethoxyphosphitomethyl[---]methylsiloxane unit is incorporated periodically in a long polydimethylsiloxane chain.

Please amend the paragraph beginning on page 15, line 8, as shown below:

A 250 ml three-necked flask with dropping funnel and reflux condenser was charged under nitrogen atmosphere with 4.9 g of diethoxyphosphitomethyldimethylmethoxysilane (0.02 mol, GC 98 %), whose preparation is described above under "Silane B". Following the addition of 0.5% by weight of a part-esterified phosphoric ester catalyst containing polyalkylene oxide units (commercially available under the brand name ARLYPON® from Cognis) and heating to 60°C, 108 g of a doubly OH-terminated polydimethylsiloxane (M = 10 800 g/mol; 0.01 mol) were slowly added dropwise over the course of 10 minutes with vigorous stirring. Subsequently the reaction mixture was heated at 80°C for 300 minutes. After the alcohol formed has been stripped off under reduced pressure, 110 g of a polydimethylsiloxane containing diethoxyphosphitomethyl end groups were obtained with an average molecular weight (GPC, number average) of 10 900 g/mol. This is a linear block copolymer in which a diethoxyphosphitomethyl[---]methylsiloxane unit is joined to each of the two ends of a polydimethylsiloxane chain.

Please amend the paragraph beginning on page 15, line 33, as shown below:

A 500 ml three-necked flask with dropping funnel and reflux condenser was charged under nitrogen atmosphere with 27.6 g of diethoxyphosphitomethyltrimethoxysilane (0.1 mol, GC 98%), whose preparation is described above under "Silane C". Following the addition of 0.5% by weight of isopropyl phosphate catalyst and heating to 60°C, 390 g of a singly OH-terminated polydimethylsiloxane (prepared by anionic addition polymerization of D3 rings, M=1300 g/mol; 0.3 mol) were slowly added dropwise over the course of 10 minutes with vigorous stirring. Subsequently the reaction mixture was heated at 80°C for 280 minutes. After the alcohol formed has been stripped off under reduced pressure, 308 g of a polydimethylsiloxane containing a diethoxyphosphitomethyl group were obtained with an average molecular weight (GPC, number average) of 4200 g/mol. This is a star-shaped block copolymer in which three polydimethylsiloxane chains are attached in the middle to a diethoxyphosphitomethyl[[]]siloxane unit.